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Manuel Gaudon

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FOLEY & LARDNER LLP
975 PAGE MILL ROAD
PALO ALTO, CA 94304

EXAMINER

EMPIE, NATHAN H

ART UNIT

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PAPER

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Office Action Summary	Application No. 10/561,581	Applicant(s) GAUDON ET AL.	
	Examiner NATHAN H. EMPIE	Art Unit 1712	

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

- 1) ☒ Responsive to communication(s) filed on 29 July 2010.
- 2a) ☐ This action is **FINAL**. 2b) ☒ This action is non-final.
- 3) ☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

- 4) ☒ Claim(s) 1-36 is/are pending in the application.
- 4a) Of the above claim(s) _____ is/are withdrawn from consideration.
- 5) ☐ Claim(s) _____ is/are allowed.
- 6) ☒ Claim(s) 1-36 is/are rejected.
- 7) ☐ Claim(s) _____ is/are objected to.
- 8) ☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

- 9) ☐ The specification is objected to by the Examiner.
- 10) ☐ The drawing(s) filed on _____ is/are: a) ☐ accepted or b) ☐ objected to by the Examiner.
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) ☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

- 12) ☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
- a) ☐ All b) ☐ Some * c) ☐ None of:
1. ☐ Certified copies of the priority documents have been received.
 2. ☐ Certified copies of the priority documents have been received in Application No. _____.
 3. ☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

* See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

- | | |
|--|---|
| 1) <input checked="" type="checkbox"/> Notice of References Cited (PTO-892) | 4) <input type="checkbox"/> Interview Summary (PTO-413)
Paper No(s)/Mail Date. _____ |
| 2) <input type="checkbox"/> Notice of Draftperson's Patent Drawing Review (PTO-948) | 5) <input type="checkbox"/> Notice of Informal Patent Application |
| 3) <input type="checkbox"/> Information Disclosure Statement(s) (PTO/SB/08)
Paper No(s)/Mail Date _____ | 6) <input type="checkbox"/> Other: _____ |

DETAILED ACTION

Continued Examination Under 37 CFR 1.114

1. A request for continued examination under 37 CFR 1.114, including the fee set forth in 37 CFR 1.17(e), was filed in this application after final rejection. Since this application is eligible for continued examination under 37 CFR 1.114, and the fee set forth in 37 CFR 1.17(e) has been timely paid, the finality of the previous Office action has been withdrawn pursuant to 37 CFR 1.114. Applicant's submission filed on 7/29/10 has been entered. Claims 1-36 are currently pending examination.

Claim Interpretation

With regard to Applicant's limitation reciting "A method of preparing a metal oxide layer on a substrate, consisting essentially of..." the Examiner asserts that the applicant's specification or claims have not provided a clear indication of what the basic and novel characteristics of the invention are. So as directed by MPEP 2111.03 [R-3]: "For the purposes of searching for and applying prior art under 35 U.S.C. 102 and 103, absent a clear indication in the specification or claims of what the basic and novel characteristics actually are, "consisting essentially of" will be construed as equivalent to "comprising." See, e.g., PPG, 156 F.3d at 1355, 48 USPQ2d at 1355".

Claim Rejections - 35 USC § 112

2. The following is a quotation of the first paragraph of 35 U.S.C. 112:

The specification shall contain a written description of the invention, and of the manner and process of making and using it, in such full, clear, concise, and exact terms as to enable any person skilled in the art to which it pertains, or with which it is most nearly connected, to make and use the same and shall set forth the best mode contemplated by the inventor of carrying out his invention.

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3. Claims 31, 32 and 36 are rejected under 35 U.S.C. 112, first paragraph, as failing to comply with the written description requirement. The claim(s) contains subject matter which was not described in the specification in such a way as to reasonably convey to one skilled in the relevant art that the inventor(s), at the time the application was filed, had possession of the claimed invention.

Claims 31 and 36 contain the limitation "wherein the suspensionB does not include an oxycarboxylic acid", this is considered a negative limitation. Applicant's specification does not explicitly direct the reader to avoid or exclude oxycarboxylic acids. "Any negative limitation or exclusionary proviso must have basis in the original disclosure. The mere absence of a positive recitation is not basis for an exclusion." MPEP 2173.05(i).

Claim 32 contain the limitation "wherein the method does not include milling", this is considered a negative limitation. Applicant's specification does not explicitly direct the reader to avoid or exclude milling processes. "Any negative limitation or exclusionary proviso must have basis in the original disclosure. The mere absence of a positive recitation is not basis for an exclusion." MPEP 2173.05(i).

Claim Rejections - 35 USC § 102

4. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

A person shall be entitled to a patent unless –

(b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

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5. Claims 1, 4, 5, 6, 8, 10, 12, 13, 19-23, 25, 27, and 33 are rejected under 35 U.S.C. 102(b) as being anticipated by Cubukcu et al (US patent 6,074,771; hereafter Cubukcu).
6. Claims 1 and 33: Cubukcu teaches a method of preparing a metal oxide layer on a substrate, (see, for example, abstract, col 11 line 45 – col14 line 37), comprising the following successive steps are carried out:
 7. a) a metal oxide powder (such as yttria stabilized zirconia, or ceria-gadolinia-zirconia) is dispersed in a liquid medium comprising a dispersion solvent (such as hexanol or hexadecane) and a dispersant (such as Witflow-910 or Sandopan MA-18), the said liquid medium containing neither plasticizer nor binder (no such additional constituents are recited), by means of which a suspension A of the said metal oxide powder in the said liquid medium is obtained (See, for example, col 11 line 45 – col 12 line 10, and col 13 lines 13 – 33);
 8. b) a solution of at least one polymer (such as poly(2-ethyhexylmethacrylate) in a solvent (such as toluene or 1-phenylnonane) is added to the said suspension A, by means of which a suspension B is obtained (See, for example, col 12 lines 10-23, and col 13 lines 33-42);
 9. c) suspension B is deposited on the substrate by a dip coating method (dipping), by means of which a green layer is obtained (See, for example, col 12 lines 24-38, and col 13 lines 42-58);
 10. d) the green layer obtained in step c) is dried (See, for example, col 12 lines 24-38, and col 13 lines 42 – 58);

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11. and e) the dried layer obtained in step d) is calcined (see, for example, heat treatment at temperatures of 100°C to 1200°C, col 12 lines 60-67, and col 13 lines 52-58).

12. Claim 4: Cubukcu further teaches the metal oxide powder as yttria stabilized zirconia or ceria-gadolinia-zirconia (See, for example, col 11 lines 45-65, and col 13 lines 7-25).

13. Claim 5: Cubukcu further teaches the metal oxide powder as cubic yttria stabilized zirconia (such as TZ8Y Tosoh powder) (see, for example, col 11 lines 45-65).

14. Claim 6: Cubukcu further teaches the dispersion solvent as hexanol hexadecane, and/or ethanol (see, for example, col 11 lines 62-67, and col 13 lines 12-42).

15. Claim 8: Cubukcu further teaches wherein the content of the metal oxide powder suspension A is 70% to 35-45% by weight (see, for example, col 13 lines 34-36).

16. Claim 10: Cubukcu further teaches wherein the dispersant comprises an ionic surfactant (see, for example, col 12 lines 13-25, such as anionic, alkyl aryl ethoxylate carboxylic acid surfactant Sandopan MA-18).

17. Claim 12: Cubukcu further teaches the mass content of the dispersant in suspension A is 0.5 % by weight relative to the mass of the dry metal oxide powder dispersed (See, for example, col 13 lines 25-33).

18. Claim 13: Cubukcu further teaches the polymer is poly(2-ethyhexylmethacrylate) (see, for example, col 12 lines 14-23).

19. Claim 19: Cubukcu further teaches the drying is carried out at a temperature of 95°C (See, for example, col 12 lines 34-38).

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20. Claim 20: Cubukcu further teaches the drying is performed over a period of time of 30 minutes (See, for example, col 12 lines 34-38).
21. Claim 21: Cubukcu further teaches the calcining step e) is carried out at a calcination temperature of 1000°C (See, for example, col 12 lines 60-68).
22. Claim 22: Cubukcu further teaches the calcination temperature is reached, starting from room temperature, at a rate of increase of 5°C / min (See, for example, col 12 lines 60-68).
23. Claim 23: Cubukcu further teaches the calcination temperature is maintained for 2 hours (see, for example, col 12 lines 60-68).
24. Claims 25 and 27: Cubukcu further teaches wherein the substrate is fully dense substrate (See for example, metal alloy, stainless steel, Ni-based alloy, aluminum alloy foils, col 10 lines 15-26, col 11 lines 45-50)

Claim Rejections - 35 USC § 103

25. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

(a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

26. Claims 2-3 and are rejected under 35 U.S.C. 103(a) as being unpatentable over Cubukcu.
27. Claims 2 and 3: Cubukcu teaches the method of claim 1 and further teaches a method of applying functional ceramic oxide coatings (such as YSZ) used in ceramic

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composite cells of electrolytic devices such as fuel cells (see, for example, abstract, col 1 line 60-col 2 line 15, Fig 1-11). Cubukcu further teaches wherein the cells (12) comprise bipolar foil (50), photolithographic foil member (60), electrical contact layer (112), preferred ceramic (such as YSZ) layer (see, for example, col 4 line 60 - col 6 line 12, Fig 2 -15). A ceramic cell is taught to have a thickness of 127 microns to 5080 microns (0.005 inches to 0.2 inches) wherein cell components such as foil 50 dimples 64 and 66 (though not explicitly the thickness of the foil itself), foil (60) and contact layer (112) together possess combined thicknesses ranging up to 0.1105 inches (2806.7 microns) leaving thicknesses for applied functional ceramic oxide layer and other components without specified thicknesses to be within the range of ~0 to 2273.3 microns. So although Cubukcu does not explicitly teach the thickness of the metal oxide layer obtained after step e) is of 1 micron to 100 microns or further 1 to 10 microns, it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a thickness within the claimed ranges since in the case where the claimed ranges "overlap or lie inside ranges disclosed by the prior art" a *prima facie* case of obviousness exists. In re Wertheim, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).

28. Claims 7, 11, 31, and 36 are rejected under 35 U.S.C. 103(a) as being unpatentable over Cubukcu as applied to claims 1, 10, and 33 above, and further in view of Mukhejee et al. ("Correlation Between Slurry Rheology, Green Density, and

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Sintered Density of Tape Cast Yttria Stabilized Zirconia” Ceramics International 27

(2001) 731-739; hereafter Mukherjee) .

29. Claims 7 and 11: Cubukcu teaches the method of claim 1 and 10 (described above) wherein Cubukcu has taught a method of applying ceramic layers (specifically YSZ), such as TOSOH TZ-8Y) via wet chemical suspension routes involving combining the ceramic powder with a dispersant in a dispersing solvent (see, for example, col 11 line 45 – col14 line 37). But Cubukcu does not explicitly teach wherein the dispersion solvent is an azeotropic mixture of ethanol and methylethyl ketone and the dispersant is a phosphate ester. Mukherjee teaches a method of making ceramic layers, specifically YSZ from TOSOH TZ-8Y powder, via wet chemical suspension routes (see, for example, abstract). Mukherjee further teaches that such YSZ suspensions can be predictably prepared by using a solvent consisting of azeotropic mixture of methyl ethyl ketone and ethanol and phosphate ester dispersant (see, for example, pg 732).

Mukherjee further teaches that phosphate ester is one of the two most popular commercial dispersants, and that it is a superior dispersant for YSZ suspensions (See, for example, pg 732 and pg 738). As both Cubukcu and Mukhejee teach methods of preparing TOSOH TZ-8Y, YSZ dispersions, it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated the azeotropic mixture of ethanol and methylethyl ketone and phosphate ester as dispersing solvent / dispersant pair in the method of Cubukcu as such a pairing it taught to be well known / popular in the art to predictably disperse TOSOH TZ-8Y, YSZ, and as such a system produces superior dispersions of such YSZ.

30. Claims 31 and 36: Cubukcu teaches the method of claims 1 and 33, and the method taught by Cubukcu in view of Mukhejee wherein an azeotropic mixture of ethanol and methylethyl ketone and phosphate ester are used dispersing solvent / dispersant pair (as taught in the rejection of claims 7 and 11, above) would result in the suspension B not including an oxycarboxylic acid.

31. Claims 9, 24, 26, and 28 are rejected under 35 U.S.C. 103(a) as being unpatentable over Cubukcu as applied to claims 1 and 27 above, and further in view of Seabaugh (US 2003/0003237; hereafter Seabaugh).

32. Claim 9: Cubukcu teaches the method of claim 1 (described above) wherein Cubukcu has taught a method of applying ceramic layers (specifically YSZ) via wet chemical suspension routes used in ceramic composite cells of electrolytic devices such as fuel cells (see, for example, abstract, col 1 line 60-col 2 line 15, Fig 1-11). Bu Cubukcu does not explicitly teach the metal oxide powder has particles the size of 5 nm to 5 micron. Seabaugh teaches it is well known in the art to apply a YSZ formed by wet chemistry methods to substrates used in solid oxide fuel cells, ceramic oxygen generation system, and ceramic membrane reactor applications (see, for example, abstract, [0002]). Seabaugh further teaches that particle size influences the film stability and has found improved coatings using bimodal distributions with YSZ particles (See, for example, [0050]), such as with sizes of ~50 nm combined with particles of 200 nm (see, for example, [0054-0055], Fig 3A). Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated the particle sizes

from about 50 – 200 nm as taught by Seabaugh into the method of Cubukcu since such sizes were found to possess improved superior coating stability.

33. Claims 24, 26, and 28: Cubukcu teach the method of claim 1 and 27, wherein Cubukcu further teaches wherein the coatings can serve in a variety of catalytic, electrolyte, and solar cell applications (see, for example, abstract, col 1 line 60-col 2 line 15, Fig 1-11). and that the coating composition is a yttrium stabilized zirconium (see, for example, col 11 lines 45-68, and col 13 lines 5-25). Seabaugh teaches a method of applying a YSZ coating via a wet chemistry method for a variety of electrochemical system applications (see, for example, abstract, [0002]).

34. Claim 24: Cubukcu does not explicitly teach wherein the metal oxide layer and the substrate undergo a co-sintering operation. Seabaugh further teaches wherein a YSZ coating layer is applied to an un-sintered ceramic substrate (see, for example, abstract, and [0062]). Seabaugh further teaches that when co-sintering the coating and substrate cracking can be avoided and dense and leak tight films can be produced (see, for example, [0014]). Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a co-sintering operation into the method of Cubukcu, as Seabaugh has taught that YSZ coating can be predictably applied to substrates via co-sintering, and since co-sintering processes help to prevent substrate / coating cracking.

35. Claim 26: Cubukcu does not explicitly teach wherein the substrate is a porous substrate ranging up to 50% by volume porosity. Seabaugh teaches it is well known in the art to apply a YSZ formed by wet chemistry method to a porous substrate of a

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ceramic electrode material, as such an articles performs well for solid oxide fuel cell, ceramic oxygen generation system, and ceramic membrane reactor applications, and that YSZ can be predictably applied to porous substrates (see, for example, [0002], and Fig 5a-c wherein the porosity of the substrate is less than 50%). As both Seabaugh and Cubukcu teach wet chemistry methods of producing YSZ coatings and Seabaugh has taught the motivation to apply YSZ coating to porous substrates to take advantage of a variety of functional applications, it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated the porous substrate such as those taught by Seabaugh (wherein the porosity of the substrate is less than 50%) into the method of Cubukcu to obtain the predictable result of forming a YSZ coating on a substrate, with the added advantage of applying the article formed to serve in solid oxide fuel cell, ceramic oxygen generation system, and ceramic membrane reactor applications.

Claim 28: Cubukcu does not explicitly teach wherein the substrate is a porous Ni-YSZ cermet substrate. Seabaugh teaches it is well known in the art to apply a YSZ formed by wet chemistry method to a porous substrate such as a of a ceramic electrode material, as such an articles performs well for solid oxide fuel cell, ceramic oxygen generation system, and ceramic membrane reactor applications, and that YSZ can be predictably applied to porous substrates such as Ni-YSZ (see, for example, [0002], [0011]). As both Seabaugh and Cubukcu teach wet chemistry methods of producing YSZ coatings and Seabaugh has taught the motivation to apply YSZ coating to porous substrates, such as Ni-YSZ to take advantage of a variety of functional applications, it

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would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated applying the YSZ coating to porous Ni-YSZ substrate as taught by Seabaugh into the method of Cubukcu as it is well known in the art to use porous Ni-YSZ cermets as substrates for YSZ coating and wherein the YSZ coated article would possess the added advantage to serve in solid oxide fuel cell application.

36. Claims 14, 30, and 35 are rejected under 35 U.S.C. 103(a) as being unpatentable over Cubukcu as applied to claims 1 and 33 above, and further in view of Valente et al (US patent 5,244,691, hereafter Valente)

37. Claims 14, 30 and 35: Cubukcu teaches the method of claim 1 and 33 (described above), but does not explicitly teaches wherein the polymer is a polymer obtainable from the reaction between hexamethylenetetramine and acetylacetone in acid medium. Valente teaches a method of depositing thin ceramic films (see, for example, abstract). Valente further teaches that it is well known in the art to use a polymer formed from a reaction between a hexamethylenetetramine and acetylacetone in an acid medium as a binding additive which aids in determining the viscosity and rheology of the coating composition (see, for example, abstract, and col 3 lines 1 – 41). Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a polymer obtainable from the reaction between hexamethylenetetramine and acetylacetone in acid medium as the polymer in the method of Cubukcu as such a polymer is well known in the art, and can predictably help to control the viscosity of the coating composition.

38. Claim 15 is rejected under 35 U.S.C. 103(a) as being unpatentable over Cubukcu as applied to claim 1 above, and further in view of Haruta et al (US 2003/0152704; hereafter Haruta).

39. Claim 15: Cubukcu teaches the method of claim 1, but does not explicitly teach wherein the solution of at least one polymer further more contains the same metals as those of the metal oxide powder. Haruta teaches a method of applying a metal oxide coating via dipcoating to a substrate surface (see, for example, abstract, [0092]). Haruta further teaches wherein the coating composition comprises a ceramic oxide (such as titanium oxide particles, see, for example, [0074], [0086]) and the addition of a halide of the same metal (such as titanium halide, which provides Ti metal, see, for example, [0080]). Haruta teaches that such an addition of metal species is well known in the art to improve coating properties such as density (see, for example [0249]-[0251]). Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have further incorporated the same metals as those of the oxide powder into the coating composition of Cubukcu, as Haruta has taught that the addition of metal species has improve coating properties such as density. Cubukcu and Haruta do not explicitly teach wherein the same metals are added specifically at the polymer solution step, but it would have been obvious to one of ordinary skill in the art at the time of invention to have added the metal species to the polymer solution since the "selection of any order of mixing ingredients is *prima facie* obvious" In re Gibson, 39 F2d 975, 5 USPQ 230 (CCPA 1930) MPEP 2144.04 IV. C.

40. Claims 16 -18, 29, and 34 are rejected under 35 U.S.C. 103(a) as being unpatentable over Cubukcu as applied to claims 1 and 33 above, and further in view of Lee ("Dip Coating of Alumina Films by the Sol-gel Method" J. Mater. Res. Vol. 8, No.12, Dec 1993, 3151-3157; hereafter Lee).

41. Claims 16-18, 29 and 34: Cubukcu teach the method of claim 1, wherein Cubukcu teaches the ceramic oxide coating is applied by dipcoating (see, for example, col 12 lines 29-38 and col 13 lines 42-52), but does not explicitly teach the viscosity of the polymer solution is 5 mPas to 1000 mPas, to combine the polymer solution and the initial oxide suspension in a mass ratio of 0.01 to 3, further 0.1 to 0.6, nor wherein the step of removing the substrate from the final suspension is at a controlled rate of 0.1 to 100 cm/min. Lee teaches a method of forming an oxide layer onto a substrate via a dipcoating method (see, for example, abstract). Lee further teaches that it is well known in the dipcoating art that factors including oxide concentration, solution viscosity, and the rate of withdrawal will influence the final coating properties such as thickness and uniformity (see, for example, pg 3154-3155). Further, one of ordinary skill in the art would appreciate that the amount of dispersant and viscosity of the polymer solution would influence the final suspension viscosity, and wherein the mass content of each suspension and solution and their mass ratio to each other would influence the final suspension's oxide concentration. Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a polymer solution viscosity of 5 mPas to 1000 mPas, to combine the polymer solution and the initial oxide

suspension in a mass ratio of 0.01 to 3, further 0.1 to 0.6 and to incorporate a controlled substrate removal rate of 0.1 to 100 cm / min since these factors are known as result effective variables, and since it has been held that where the general conditions of a claim are disclosed in the prior art, discovering the optimum or workable ranges involves only routine skill in the art. In re Aller, 220 F.2d 454, 456, 105 USPQ 233, 235 (CCPA 1955).

42. Claims 32 is rejected under 35 U.S.C. 103(a) as being unpatentable over Cubukcu as applied to claim 1 above, and further in view of Kuno et al (US patent 6,221,945; hereafter Kuno).

43. Claim 32: Cubukcu teaches the method of claim 1 (described above), wherein Cubukcu has taught preparing the dispersion in an attritor mill (see, for example, col 11 lines 55-62), so Cubukcu does not explicitly teach wherein the method does not include milling. Kuno teaches a method of preparing metal oxide containing dispersions for coatings (See, for example, abstract, col 3 lines 1-36). Kuno further teaches wherein the dispersions are well known in the art to be prepared by mills or by an ultrasonic disperser (See, for example, col 4 lines 33-36). As both Cubukcu and Kuno teach method involving dispersing metal oxide containing dispersions, it would have been obvious to one of ordinary skill in the art at the time of invention to substitute one method for the other (ultrasonic dispersing for milling) to achieve the predictable result of preparing a dispersion comprising ceramic oxide particles.

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44. Claims 1-6, 9-10, 13, 19, 21-23, 25, 27, 31-33 and 36 are rejected under 35 U.S.C. 103(a) as being unpatentable over Nonninger et al (WO 02/086194, as supplied by applicant's IDS, the examiner will be making references in regard to English equivalent US 2004/0115416 A1; hereafter Nonninger), in view of Bitterlich et al.

("Rheological Characterization of Water-Based Slurries for the Tape Casting Process" Ceramics International 28 (2002) 675-683).

45. Claims 1 and 33: Nonninger teaches a method of preparing a metal oxide layer on a substrate (see, for example, abstract), comprising the following successive steps of:

46. dispersing a metal oxide powder (see, for example, [0012-0013], preferably a ysz is taught) in a liquid medium comprising a dispersion solvent (see, for example, [0020], such as water) and a dispersant (see, for example, [0019]), the said liquid medium containing neither plasticizer nor binder, by means of which a suspension A of the said metal oxide powder in the said liquid medium is obtained (see, for example, in [0029] wherein YSZ particles, and a dispersant are first mixed prior to subsequent polymer additive additions);

47. adding at least one polymer to the suspension A, obtaining suspension B (see, for example, further polymeric additives ([0016], [0029], [0031]) being added after mixing suspension A (see, for example, [0029], [0031], and claim 1 wherein the order of adding the polymer / solvent mixture is taught subsequent to preparing suspension A).

48. depositing suspension B on the substrate by a dip coating method by means of which a green layer is obtained (see, for example, [0009], [0011], [0029], [0031]),

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49. the green layer is dried and calcined to obtain said metal oxide layer on said

substrate (see, for example, [0011], [0029], [0031])

50. Nonninger further teaches wherein suspension A is mixed with a mixture of at

least one solvent and at least one polymer, but does not explicitly teach wherein the mixture is a solution. Bitterlich teaches a method of making stable ceramic suspensions

(specifically for t-YSZ, pg 676) for coating processes (see, for example, abstract, pg 675-676), which further involve a two step mixing process wherein the powder / solvent / dispersant are first prepared and mixed, and then solution of a polymer (binder) / solvent are added to the first mixture (see, for example, pg 676-677). Bitterlich teaches that it is well known in the art to provide a binder with a solvent to predictably add a binder to suspension (see, for example, pg 676-677). As both Nonninger and Bitterlich

teach methods wherein a polymeric / binder addition is applied following dispersant addition, and as both are directed to methods of forming films of ceramic (specifically yttrium stabilized zirconia) it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated the polymer / binder in a solution with a solvent as the means to introduce the polymer / binder in the method of Nonninger, as

Bitterlich has taught that polymeric additives can predictably be incorporated into dispersions with a solvent in the form of a solution, and as one of ordinary skill in the art would appreciate that additions of solvent can be used to control the viscosity and mixing behavior of the suspension,

51. Claims 2 and 3: Nonninger further teaches wherein the oxide layer has a thickness of between 100 nm and 10 microns (see, for example, [0023]). Although

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Deleted: Mukherjee teaches a method of making ceramic layers (specifically YSZ) via wet chemical suspension routes (see, for example, abstract). Mukherjee further teaches that it is well known in the art that the sequence of adding additives to a suspension is critical, namely the dispersant has to be added before the other polymeric additives properly break down any agglomerates generating a proper dispersion (pg 732). Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have added the polymer / binder species subsequent to the dispersant, as taught by Mukherjee, to avoid competitive adsorption and break down any agglomerates in the method of Nonninger.

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Nonninger does not explicitly teach the oxide layer having a thickness of 1 to 100 microns, it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated such a claimed thickness since in the case where the claimed ranges “overlap or lie inside ranges disclosed by the prior art” a *prima facie* case of obviousness exists. In re Wertheim, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).

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52. Claim 4: Nonninger further teaches a variety of metal oxides, including simple oxides of transition metals and lanthanides (see, for example, [0012])

53. Claim 5: Nonninger further teaches wherein the metal oxide is preferably an yttrium stabilized zirconia ([0013]) and Bitterlich further explicitly teaches a YSZ with a cubic structure (see, for example TZ8Y, pg 676 section 2.1).

54. Claim 6: Nonninger further explicitly teaches the dispersion solvent as water (see, for example, [0020])

55. Claim 9: Nonninger further teaches wherein the metal oxide powder particles have a size of between 3 nm and 100 nm (see, for example, [0026]); although Nonninger does not explicitly teach 5 nm to 5 microns; it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated such a claimed size since in the case where the claimed ranges “overlap or lie inside ranges disclosed by the prior art” a *prima facie* case of obviousness exists. In re Wertheim, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).

56. Claims 10: Nonninger further teaches wherein the dispersant is chosen from ionic and non-ionic surfactants (surface active molecules) (see, for example, [0019]).

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57. Claim 13: Nonninger further teaches wherein the polymer is chosen from poly(aliphatic) esters (see, for example, polyester, [0016]).
58. Claim 19: Nonninger further teaches wherein the drying is conducted at 80°C. (see, for example [0029]).
59. Claim 21: Nonninger further teaches wherein the calcination step is conducted at 500°C ([0029]). And alternatively, Bitterlich further teaches calcining YSZ at 650°C, 1200°C (see, for example, pg 677).
60. Claim 22: Nonninger further teaches wherein the heating rate 5°C / min (5 K / min) (see, for example, [0029]). And alternatively, Bitterlich further teaches firing YSZ at heating rates of 25°C / min and 60°C /min (see, for example, pg 677).
61. Claim 23: Nonninger further teaches wherein the calcination temperature is held for 1 hour (see, for example, [0029]). Alternatively Bitterlich further teaches a hold for 3 hours (see, for example, pg 677).
62. Claims 25 and 27: Nonninger further teaches wherein the substrate is a fully dense substrate (see, for example, steel substrate, [0029]). Formatted: Font:
63. Claims 31 and 36: Nonninger further teaches wherein the suspension B does not include an oxycarboxylic acid (See, for example, wherein the surface active molecule is a beta di-ketone, or alkylsilane, which are not oxycarboxylic acid, [0019]). Formatted: Font:
64. Claim 32 Nonninger in view of Bitterlich teach the method of claim 1, and the process of Nonninger does not explicitly recite any milling steps, so it would not include milling.

65. Claims 7 and 11 are rejected under 35 U.S.C. 103(a) as being unpatentable over Nonninger in view of Bitterlich as applied to claims 1 and 10 above, and further in view of Mukherjee.

66. Claims 7 and 11: Nonninger in view of Bitterlich teaches the method of claim 1 and 10 (described above) wherein Nonninger has taught a method of applying ceramic layers (specifically YSZ), via wet chemical suspension routes involving combining the ceramic powder with a dispersant in a dispersing solvent (see, for example, [0012],[0019-0020], [0029], [0031]). But Nonninger does not explicitly teach wherein the dispersion solvent is an azeotropic mixture of ethanol and methylethyl ketone and the dispersant is a phosphate ester. Mukherjee teaches a method of making ceramic layers, specifically YSZ powder, via wet chemical suspension routes (see, for example, abstract). Mukherjee further teaches that such YSZ suspensions can be predictably prepared by using a solvent consisting of azeotropic mixture of methyl ethyl ketone and ethanol and phosphate ester dispersant (see, for example, pg 732). Mukherjee further teaches that phosphate ester is one of the two most popular commercial dispersants, and that it is a superior dispersant for YSZ suspensions (See, for example, pg 732 and pg 738). As both Mukherjee and Nonninger in view of Bitterlich teach methods of preparing YSZ dispersions comprising dispersants and dispersing solvents it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated the azeotropic mixture of ethanol and methylethyl ketone and phosphate ester as dispersing solvent / dispersant pair in the method of Cubukcu as such a pairing

it taught to be well known / popular in the art to predictably disperse YSZ powders, and as such a system produces superior dispersions of such YSZ.

67. Claim 20 is rejected under 35 U.S.C. 103(a) as being unpatentable over Nonninger in view of Bitterlich as applied to claim 19 above, and further in view of Applicant's admitted prior art (AAPA).

68. Claim 20: Neither of Nonninger, or Bitterlich explicitly teach the duration of drying. It is well known in the art that the duration of drying is a result effective variable, influencing a balance between shorter processing times and complete drying (AAPA) and as such it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a drying time of from 1 min to 10 hrs since it has been held that where the general conditions of a claim are disclosed in the prior art, discovering the optimum or workable ranges involves only routine skill in the art. In re Aller, 220 F.2d 454, 456, 105 USPQ 233, 235 (CCPA 1955).

69. Claims 14, 30, and 35 are rejected under 35 U.S.C. 103(a) as being unpatentable over Nonninger in view of Bitterlich as applied to claims 1 and 33 above, and further in view of Valente.

70. Claims 14, 30, and 35: Nonninger in view of Bitterlich teach the method of claims 1 and 33 (described above), but none explicitly teaches wherein the polymer is a polymer obtainable from the reaction between hexamethylenetetramine and acetylacetone in acid medium. Valente teaches a method of depositing thin ceramic

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films (see, for example, abstract). Valente further teaches that it is well known in the art to use a polymer formed from a reaction between a hexamethylenetetramine and acetylacetone in an acid medium as a binding additive which aids in determining the viscosity and rheology of the coating composition (see, for example, abstract, and col 3 lines 1 – 41). Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a polymer obtainable from the reaction between hexamethylenetetramine and acetylacetone in acid medium as the polymer in the method of Nonninger in view of Bitterlich as such a polymer is well known in the art, and can predictably help to control the viscosity of the coating composition.

71. Claim 15 is rejected under 35 U.S.C. 103(a) as being unpatentable over Nonninger in view of Bitterlich as applied to claim 1 above, and further in view of Haruta.

72. Nonninger in view of Bitterlich teach the method of claim 1, but none explicitly teach wherein the solution of at least one polymer further more contains the same metals as those of the metal oxide powder. Haruta teaches a method of applying a metal oxide coating via dipcoating to a substrate surface (see, for example, abstract, [0092]). Haruta further teaches wherein the coating composition comprises a ceramic oxide (such as titanium oxide particles, see, for example, [0074], [0086]) and the addition of a halide of the same metal (such as titanium halide, which provides Ti metal, see, for example, [0080]). Haruta teaches that such an addition of metal species is well known in the art to improve coating properties such as density (see, for example [0249]-[0251]). Therefore it would have been obvious to one of ordinary skill in the art at the

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time of invention to have further incorporated the same metals as those of the oxide powder into the coating composition of Nonninger in view of Bitterlich, as Haruta has taught that the addition of metal species has improve coating properties such as density. Nonninger in view of Bitterlich and Haruta do not explicitly teach wherein the same metals are added specifically at the polymer solution step, but it would have been obvious to one of ordinary skill in the art at the time of invention to have added the metal species to the polymer solution since the "selection of any order of mixing ingredients is *prima facie* obvious" In re Gibson, 39 F2d 975, 5 USPQ 230 (CCPA 1930) MPEP 2144.04 IV. C.

73. Claims 8, 12, 16 -18, 29 and 34 are rejected under 35 U.S.C. 103(a) as being unpatentable over Nonninger in view of Bitterlich as applied to claims 1 and 33 above, and further in view of Lee.

74. Claims 8, 12, 16-18, 29 and 34: Nonninger in view of Bitterlich teach the method of claims 1 and 33 (described above), wherein Nonninger teaches the coating is applied by dipcoating ([0009]), but none explicitly teach wherein the metal oxide powder content in the initial oxide suspension of 1 to 80%, the mass content of dispersant in the initial oxide suspension of from 0.1 to 10% by weight relative to the mass of dry metal oxide powder dispersed, the viscosity of the polymer solution is 5 mPas to 1000 mPas, to combine the polymer solution and the initial oxide suspension in a mass ratio of 0.01 to 3, further 0.1 to 0.6, nor wherein the step of removing the substrate from the final suspension is at a controlled rate of 0.1 to 100 cm/min. Lee teaches a method of

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forming an oxide layer onto a substrate via a dipcoating method (see, for example, abstract). Lee further teaches that it is well known in the dipcoating art that factors including oxide concentration, solution viscosity, and the rate of withdrawal will influence the final coating properties such as thickness and uniformity (see, for example, pg 3154-3155). Further, one of ordinary skill in the art would appreciate that the amount of dispersant and viscosity of the polymer solution would influence the final suspension viscosity, and wherein the mass content of each suspension and solution and their mass ratio to each other would influence the final suspension's oxide concentration. Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a metal oxide powder content in the initial ceramic suspension of 1 to 80%, a mass content of dispersant in the initial ceramic suspension of from 0.1 to 10% by weight relative to the mass of dry metal oxide powder added, a polymer solution viscosity of 5 mPas to 1000 mPas, to combine the polymer solution and the initial oxide suspension in a mass ratio of 0.01 to 3, further 0.1 to 0.6, and to incorporate a controlled substrate removal rate of 0.1 to 100 cm / min since these factors are known as result effective variables, and since it has been held that where the general conditions of a claim are disclosed in the prior art, discovering the optimum or workable ranges involves only routine skill in the art. In re Aller, 220 F.2d 454, 456, 105 USPQ 233, 235 (CCPA 1955).

75. Claims 24, 26, and 28 are rejected under 35 U.S.C. 103(a) as being unpatentable over Nonninger in view of Bitterlich as applied to claim 1 above, and further in view of Seabaugh.

76. Claims 24, 26, and 28: Nonninger in view of Bitterlich teach the method of claim 1 and 27, wherein Nonninger further teaches the substrate as metal, glass, enamel or ceramic (see, for example [0023]). Nonninger further teaches wherein the coatings can serve in a variety of catalytic, electrolyte, and solar cell applications ([0002]), and that the coating composition is preferably a yttrium stabilized zirconium ([0013]). Seabaugh teaches a method of applying a YSZ coating via a wet chemistry method for a variety of electrochemical system applications (see, for example, abstract, [0002]).

77. Claim 24: Nonninger in view of Bitterlich do not explicitly teach wherein the metal oxide layer and the substrate undergo a co-sintering operation. Seabaugh further teaches wherein a YSZ coating layer is applied to an un-sintered ceramic substrate (see, for example, abstract, and [0062]). Seabaugh further teaches that when co-sintering the coating and substrate cracking can be avoided and dense and leak tight films can be produced (see, for example, [0014]). Therefore it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a co-sintering operation into the method of Nonninger in view of Bitterlich, as Seabaugh has taught that YSZ coating can be predictably applied to substrates via co-sintering, and since co-sintering processes help to prevent substrate / coating cracking.

78. Claim 26: Nonninger in view of Bitterlich do not explicitly teach wherein the substrate is a porous substrate ranging up to 50% by volume porosity. Seabaugh

teaches it is well known in the art to apply a YSZ formed by wet chemistry method to a porous substrate of a ceramic electrode material, as such an articles performs well for solid oxide fuel cell, ceramic oxygen generation system, and ceramic membrane reactor applications, and that YSZ can be predictably applied to porous substrates (see, for example, [0002], and Fig 5a-c wherein the porosity of the substrate is less than 50%).

As both Seabaugh and Nonninger in view of and Bitterlich teach wet chemistry methods of producing YSZ coatings and Seabaugh has taught the motivation to apply YSZ coating to porous substrates to take advantage of a variety of functional applications, it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated the porous substrate such as those taught by Seabaugh (wherein the porosity of the substrate is less than 50%) into the method of Nonninger in view of Bitterlich to obtain the predictable result of forming a YSZ coating on a substrate, with the added advantage of applying the article formed to serve in solid oxide fuel cell, ceramic oxygen generation system, and ceramic membrane reactor applications.

79. Claim 28: Nonninger in view of Bitterlich do not explicitly teach wherein the substrate is a porous Ni-YSZ cermet substrate. Seabaugh teaches it is well known in the art to apply a YSZ formed by wet chemistry method to a porous substrate such as a of a ceramic electrode material, as such an articles performs well for solid oxide fuel cell, ceramic oxygen generation system, and ceramic membrane reactor applications, and that YSZ can be predictably applied to porous substrates such as Ni-YSZ (see, for example, [0002], [0011]). As both Seabaugh and Nonninger in view of Bitterlich teach wet chemistry methods of producing YSZ coatings and Seabaugh has taught the

motivation to apply YSZ coating to porous substrates, such as Ni-YSZ to take advantage of a variety of functional applications, it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated applying the YSZ coating to porous Ni-YSZ substrate as taught by Seabaugh into the method of Nonninger in view Bitterlich as it is well known in the art to use porous Ni-YSZ cermets as substrates for YSZ coating and wherein the YSZ coated article would possess the added advantage to serve in solid oxide fuel cell applications.

Response to Arguments

80. Applicant's arguments filed 7/29/10 have been fully considered but they are not persuasive.

81. Upon reconsideration of Nonninger reference, particularly ([0029], [0031] and claim 1, wherein Nonninger has clearly taught the claimed order of addition of a mixture of a polymer with a solvent to a dispersion of mixed oxide powder the examiner has no longer relied upon the teaching of secondary reference Mukherjee to reject claims 1-6, 9-10, 13, 19, 21-23, 25, 27, 31-33 and 36.

82. Additionally examiner's further searching has yielded the Cubukcu reference, which has been applied against the recited claims alone and in combination with the appropriate secondary references, as described in the rejection above.

83. A. In response to applicant's arguments "Bitterlich does not teach a polymer solution" (pg 10-11), the examiner directs the applicant's attention to applicant's own specification (see, for example, pg 13 line 30 - pg 15 line 8) it would appear that a sol is considered as a "solution". The examiner asserts that a sol is well known in the art as a

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liquid colloidal dispersion. Further by applicant's omission "the term 'sol', when used in the context of organic chemistry can refer to a solution." (pg 10 of remarks dated 1/29/10). According to Bitterlich: "The emulsion binders, which are colloidal dispersions of a polymer in water..." (pg 675 1st paragraph). As the applicant's own invention encompasses polymeric sols as solutions, and as the Applicant has agreed that sol can refer to a solution, the prior art should be privy to just such a scope, and as such presents sufficient teaching to a medium that would be considered a solution by applicant's standards. The examiner asserts that the binders of Bitterlich are organic thus would fall within the context of organic chemistry so they would too satisfy the limitation to a solution. Additionally, the examiner had previously cited prior art references Graham et al (US 2005/010697): "The term 'solution', as defined herein is understood to include liquids systems, gaseous systems, gels, suspensions, colloids, slurries, emulsions, and the like, and mixtures of any two or more or the forgoing" ([0038]); and Scholtz et al. (US patent 5,723,174) "The term 'solution' as used herein includes dispersion or suspensions of finely divided inorganic metal oxide particles in a liquid medium" (col 5 lines 14 - 16); to further support that the art has recognized emulsions, and colloidal dispersions as solutions; and therefore the cited prior art has sufficiently taught polymer solutions.

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84. B / D. In response to applicant's arguments directed to milling taught by Mukherjee reference (pg 11-13 of remarks), the examiner asserts that the Mukherjee reference is no longer relied upon to reject claim 1 from which newly dependant claim 31 directed to milling depends from. Therefore the applicant's arguments are moot.

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Furthermore, the milling steps of Mukherjee / Bittlerling were never relied upon in the rejection, and the relied upon process of primary reference Nonninger has not taught milling. The test for obviousness is not whether the features of a secondary reference may be bodily incorporated into the structure of the primary reference; nor is it that the claimed invention must be expressly suggested in any one or all of the references.

Rather, the test is what the combined teachings of the references would have suggested to those of ordinary skill in the art. See *In re Keller*, 642 F.2d 413, 208 USPQ 871 (CCPA 1981).

85. C. In response to applicant's argument that "Nonninger requires an oxycarboxylic acid" (pg 12 of remarks) the examiner disagrees with applicant's position. Nonninger has taught oxycarboxylic acid as the surface active molecules (dispersant), but Nonninger has additionally taught other non-oxycarboxylic acid dispersants can be used instead, such as alkylsilanes which are not oxycarboxylic acid. Therefore it would be proper to conduct the invention of Nonninger without a oxycarboxylic acid because such embodiments are explicitly taught by Nonninger.

86. E. In response to applicant's arguments that the prior art do not teach the newly added claims, the examiner directs the applicant's attention to the rejections of such claims above. Further with respect to the applicant's argued claimed r_m ratio, the examiner asserts that such new limitation is just a narrower range to previously recited r_m range of claim 17, so it was rejected similarly as claim 17.

87. As to the dependent claims, they remain rejected as no separate arguments are provided.

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As the applicant has not traversed the examiners previous assertion of official notice "It is well known in the art that the duration of drying is a result effective variable, influencing a balance between shorter processing times and complete drying"(refer to content rejecting claim 20 in previous office action), the well known in the art statement is taken to be admitted prior art (AAPA).

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Conclusion

Any inquiry concerning this communication or earlier communications from the examiner should be directed to NATHAN H. EMPIE whose telephone number is (571)270-1886. The examiner can normally be reached on M-F, 6:45- 4:15 EST.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Michael Cleveland can be reached on (571) 272-1418. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

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Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Nathan H Empie/
Examiner, Art Unit 1712

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Claim 5: Nonninger further teaches wherein the metal oxide is preferably an yttrium stabilized zirconia ([0013]) and Bitterlich further explicitly teaches a YSZ with a cubic structure (see, for example TZ8Y, pg 676 section 2.1).¶
 Claim 6: Nonninger further explicitly teaches the dispersion solvent as water (see, for example, [0020])¶
 Claim 7: Mukherjee explicitly teaches a preferred solvent for a YSZ suspension as an azeotropic mixture of methyl ethyl ketone and ethanol (pg 732).¶
 Claim 9: Nonninger further teaches wherein the metal oxide powder particles have a size of between 3 nm and 100 nm (see, for example, [0026]); although Nonninger does not explicitly teach 5 nm to 5 microns; it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated such a claimed size since in the case where the claimed ranges "overlap or lie inside ranges disclosed by the prior art" a *prima facie* case of obviousness exists. In re Wertheim, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).¶
 Claims 10 and 11: Nonninger further teaches wherein the dispersant is chosen from ionic and non-ionic surfactants (surface active molecules) (see, for example, [0019]).
 Additionally, Mukherjee explicitly teaches that phosphate ester is a popular dispersant for YSZ suspensions (pg 732).¶
 Claim 13: Nonninger further teaches wherein the polymer is chosen from poly(aliphatic) esters (see, for example, polyester, [0016]).¶
 Claim 19: Nonninger further teaches wherein the drying is conducted at 80°C. (see, for example [0029]).¶
 Claim 20: None of Nonninger, Mukherjee, or Bitterlich explicitly teach the duration of drying. The examiner takes official notice that it is well known in the art that the duration of drying is a result effective variable, influencing a balance between shorter processing times and complete drying and as such it would have been obvious to one of ordinary skill in the art at the time of invention to have incorporated a drying time of from 1 min to 10 hrs since it has been held that where the general conditions of a claim are disclosed in the prior art, discovering the optimum or workable ranges involves only routine skill in the art. In re Aller, 220 F.2d 454, 456, 105 USPQ 233, 235 (CCPA 1955).¶
 Claim 21: Nonninger further tea ... [1]